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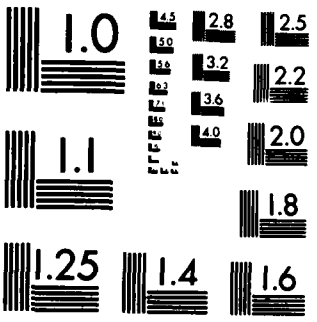

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The mechanism of the enhancement of divacancy production by oxygen during electron irradiation of silicon. II. Computer modeling

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I. INTRODUCTION

In a previous paper (Part I) we reported experimental results about the influence of the interstitial oxygen content of a Czochralski (Cz) silicon wafer on the divacancy production during room-temperature electron irradiation.[1] We found that the divacancy [V_2] introduction rate varied linearly with interstitial oxygen concentration. Similarly the introduction rate of vacancy-oxygen centers (VO) displayed a linear dependence on interstitial oxygen content. The most remarkable result of our experiment was that the divacancy concentration correlated much better to the density of introduced vacancy-oxygen centers than to the initially present dissolved oxygen concentration and that the relationship between V_2 and VO was perfectly linear

The VO density of a Si sample after electron irradiation is an indicator of how many single vacancies were available for capture by the oxygen atoms, i.e., the VO concentration is proportional to the steady-state concentration of vacancies and the length of the irradiation (neglecting saturation and other effects). The linear relationship between V_2 and VO indicates a possible dependence of the V_2 concentration on the steady-state vacancy concentration, i.e., the importance of the indirect production of divacancies.

Based on our results we proposed different possible models for the increase in divacancy introduction rate with increasing oxygen content. It is the purpose of the present paper to test these models numerically. Of the possible divacancy enhancement mechanisms mentioned in Part I only one can reproduce all our experimental results.

II. DEFECT PRODUCTION MODELING

A. Basic model

In our computer model of defect production[2-4] we assume that single vacancies V and Si self-interstitials I are created at a constant rate G_0 during room-temperature electron bombardment of silicon. Simultaneously the irradiation introduces immobile divacancies V_2 and a corresponding number of interstitials at a constant rate G_{V_2O}. The mobile interstitials and vacancies diffuse and can annihilate each other or they can be captured by trapping centers. In addition two vacancies are allowed to combine and form a divacancy. The capture rate between any two defects is assumed to be of the form $4\pi(D_1 + D_2)R_{12}$, where D_1 and D_2 are the diffusion coefficients of the defects and R_{12} is an effective capture radius.[5,6] For the boron-doped Cz silicon crystal ($\rho \approx 7\Omega$ cm) which we used in our experiment[1] we restrict ourselves to these impurities: Varying interstitial oxygen concentrations, a substitutional carbon density of 5.0×10^{16} at/cm^3, and a substitutional boron content of 2.0×10^{15} at/cm^3.[1]

It is known that the interstitial oxygen O_I captures vacancies to form vacancy-oxygen centers VO,[7] while the substitutional carbon C_s and boron B_s impurities trap Si self-interstitials.[8,9] Interstitial carbon C_I is mobile at room temperature and can react with other defects. Interstitial boron B_I complexes with other Si self-interstitials to form large interstitial clusters $B_I I_n$.[10]

In the simplest version of our model the following defect reactions are assumed:

$$V + I \rightarrow \phi \text{ (recombination)}, \quad (1)$$
$$V + V \rightarrow V_2, \quad (2)$$
$$V_2 + I \rightarrow V, \quad (3)$$
$$V + O_I \rightarrow VO, \quad (4)$$
$$VO + I \rightarrow O_I, \quad (5)$$
$$I + C_s \rightarrow C_I, \quad (6)$$
$$C_I + V \rightarrow C_s, \quad (7)$$
$$VO + C_I \rightarrow C_s O_I, \quad (8)$$
$$B_s + I \rightarrow B_I, \quad (9)$$
$$B_I + I \rightarrow B_I I, \quad (10)$$
$$B_I I + I \rightarrow B_I I_2, \quad (11)$$
$$\vdots$$
$$B_I I_n + I \rightarrow B_I I_{n+1}.$$

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TABLE I. Simultaneous differential equations for the reactions (1–11).

$$\frac{\partial[V]}{\partial t} = G_0 - R'(V,I)(D_I + D_V)[V][I] - 2R'(V,V)D_V[V]^2 - R'(V,O_I)D_V[V][O_I] - R'(C_I,V)(D_{C_I} + D_V)[C_I][V] + R'(V_2,I)D_I[V_2][I]$$

$$\frac{\partial[I]}{\partial t} = G_0 + 2G_{V_2O} - R'(V,I)(D_I + D_V)[V][I] - R'(VO,I)D_I[VO][I] - R'(V_2,I)D_I[V_2][I] - R'(C_S,I)D_I[C_S][I] - R'(B_I,I)[B_I][I]$$

$$\frac{\partial[V_2]}{\partial t} = G_{V_2O} + 2R'(V,V)D_V[V]^2 - R'(V_2,I)D_I[V_2][I]$$

$$\frac{\partial[O_I]}{\partial t} = -R'(O_I,V)D_V[O_I][V] + R'(VO,I)D_I[VO][I]$$

$$\frac{\partial[VO]}{\partial t} = R'(O_I,V)D_V[O_I][V] - R'(VO,I)D_I[VO][I] - R'(C_I,VO)D_{C_I}[C_I][VO]$$

$$\frac{\partial[C_S]}{\partial t} = -R'(C_S,I)D_I[C_S][I] + R'(C_I,V)(D_{C_I} + D_V)[C_I][V]$$

$$\frac{\partial[C_I]}{\partial t} = R'(C_S,I)D_I[C_S][I] - R'(C_I,V)(D_{C_I} + D_V)[C_I][V] - R'(C_I,VO)D_{C_I}[C_I][VO]$$

$$\frac{\partial[B_S]}{\partial t} = -R'(B_S,I)D_I[B_S][I]$$

$$\frac{\partial[B_I]}{\partial t} = R'(B_S,I)D_I[B_S][I]$$

always $R'=(X,Y)=4\pi R(X,Y)$.

These reactions require the system of simultaneous differential equations shown in Table I.[11] We solved this system numerically by using a computer. Values for the various physical parameters which we used in the calculation are shown in Table II.

All capture radii are taken to be 5 Å which is appropriate for neutral defects and elastic interaction at 300 K.[12] We neglect charge state effects, although they could be important.[12] The values of the diffusion coefficients for the different species which are shown in Table II are based mainly on experimental values for the migration energy and the assumption that the jump frequency is equal to the Debye frequency.[13] Our confidence in D_v is good.[13–17] However, we are much less certain about the accuracy of D_I,[16,17] D_{C_I},[18] and D_{B_I}.[19] Ionization effects on the diffusion of the defects are neglected in this paper. The vacancy-interstitial generation rate shown in Table II is converted into fluence rate by assuming that the number of V, I pairs produced per 2-MeV electron is 0.5[20] and that only 10% of all V, I pairs escape correlated recovery. Table II also shows the initial concentrations of the various defects.

TABLE II. Input parameters used in numerical computations.

Capture radii: $R(X,Y) = 5$ Å for all defects X and Y.

Diffusion coefficients (300 K):

$D_I = 3.16\times10^{-4}$ cm²/s,
$D_V = 4.15\times10^{-9}$ cm²/s,
$D_{C_I} = 1.0\times10^{-14}$ cm²/s,
$D_{B_I} = 1.0\times10^{-12}$ cm²/s.

Generation rates:

$G_0 = 5.0\times10^{16}$ cm⁻³/s,
$G_{V_2O} = 2.0\times10^{16}$ cm⁻³/s.

Initial values of defect concentrations:

$[O_I]$ = varying: 5.0×10^{16} atoms/cm³ to 1.0×10^{18} atoms/cm³,
$[C_S] = 5.0\times10^{16}$ atoms/cm³,
$[B_S] = 2.0\times10^{15}$ atoms/cm³.

All others = 0.

B. Application to the question of the role of oxygen in divacancy production

In Part I we proposed different possible mechanisms for the oxygen dependence of the divacancy production rate in electron-irradiated silicon at room temperature. We tested those suggestions numerically with our computer model of radiation damage. Below we list the different mechanisms mentioned in Part I and the necessary modifications to the model described in Sec. II A.

1. Kinetics

Only the solution of the system shown in Table I is required.

2. Brelot's Suggestion[21]

We have to include the capture of Si self-interstitials by interstitial oxygen and the possible interactions of the resulting complex with other defects. The following defect reactions were added to Eqs. (1)–(11):

$$O_I + I \rightarrow O_I I, \tag{12}$$

$$O_I I + V \rightarrow O_I. \tag{13}$$

3. V_2O Center

We allow here for the formation of a V_2O complex from VO. The respective concentrations of V_2O and V_2 are added to get an "apparent" divacancy concentration. We added the following reactions to Eqs. (1)–(11):

$$V + VO \rightarrow V_2O, \tag{14}$$

$$V_2O + I \rightarrow VO. \tag{15}$$

4. Impurity dependence of primary damage

a. Generation rate of V, I pairs. The introduction rate of V, I pairs is assumed to depend on the interstitial oxygen content. We therefore write

$$G(O_I) = G_0 + C[O_I]. \tag{16}$$

b. Direct generation rate of divacancies. We allow the direct generation rate of V_2 to depend on the interstitial oxygen concentration. The constant G_{V_2} is therefore replaced by

$$G_{V_2}(O_I) = G_{V_2O} + C[O_I]. \tag{17}$$

For both cases (*a* and *b*) we used $C = 0.1$ s⁻¹.

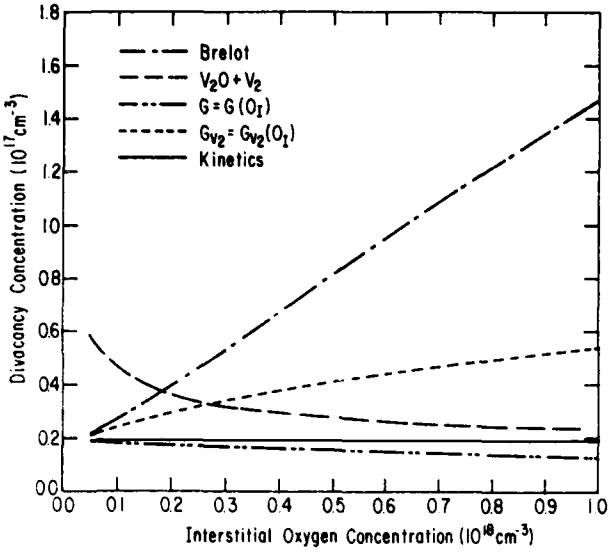

FIG. 1. Numerical results for the divacancy concentrations vs increasing initial oxygen concentration after a total electron dose of 1.0×10^{18} electrons/cm^2. "Brelot," "$V_2O + V_2$," "$G = G(O_I)$," "$G_{V_2} = G_{V_2}(O_I)$," and "Kinetics" denote the models described in Ref. [1] and in Sec. II B.

III. RESULTS

In Fig. 1 we show the divacancy concentrations which we obtain after a dose of 1.0×10^{18} electrons/cm^2 for the different models described in Sec. II B versus varying initial interstitial oxygen contents. Only Brelot's model and $G_{V_2}(O_I)$ give increasing $[V_2]$ versus increasing $[O_I]$. All other models give either nearly constant $[V_2]$ (kinetics, i.e., the simplest system) or decreasing $[V_2]$ versus increasing interstitial oxygen $[V_2O + V_2$ or $G(O_I)]$.

Figure 2 shows the vacancy-oxygen concentrations obtained in the same calculations. All five models show an increasing A-center density versus rising initial amounts of oxygen. In Brelot's model there exists a nearly perfect linear relationship between [VO] and $[O_I]$ over the whole range of $[O_I]$ values. This is the case for the other models only for larger $[O_I]$.

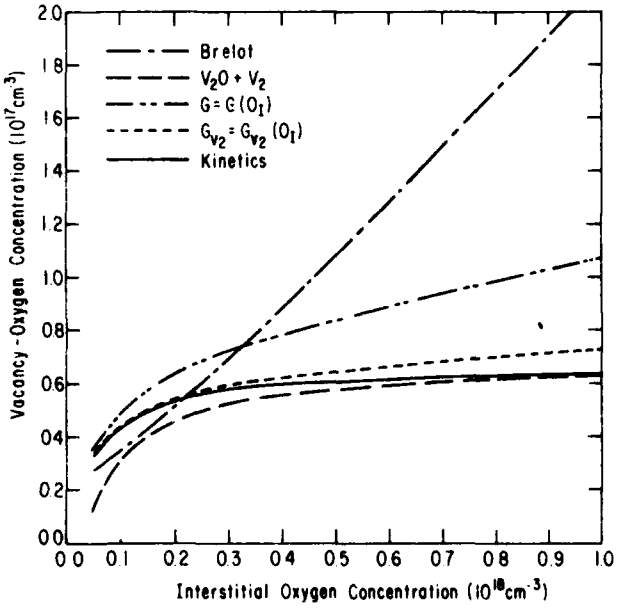

FIG. 2. Numerical results for the vacancy-oxygen concentrations vs increasing initial oxygen concentration obtained in the same calculations.

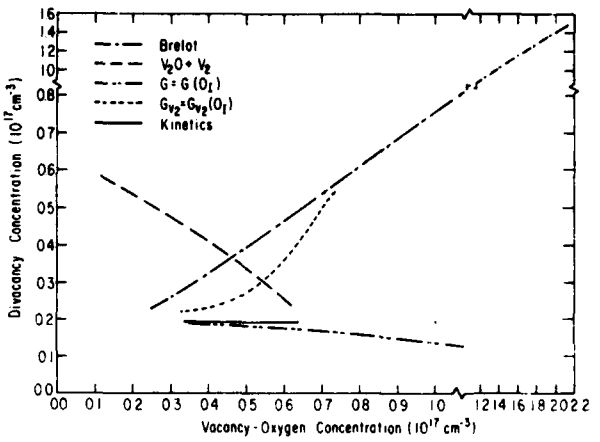

FIG. 3. The numerical data of Figs. 1 and 2, but here plotting divacancy concentration vs the corresponding vacancy-oxygen concentration.

The relationship between $[V_2]$ and [VO] after an electron dose of 1.0×10^{18} electrons/cm^2 is shown in Fig. 3. The models $V_2O + V_2$, $G(O_I)$ and the simplest system (kinetics) all show declining divacancy concentrations with larger vacancy-oxygen concentrations. In Brelot's model $[V_2]$ rises nearly linearly with [VO], while for $G_{V_2}(O_I)$ the divacancy concentration is also increasing, but the relationship to [VO] is not linear.

These qualitative trends of the different models are stable with respect to variation of the input parameters, i.e., diffusion coefficients, capture radii, and C (in Eqs. 16 and 17), within reasonable limits. We therefore believe that the results displayed in Figs. 1–3 reflect intrinsic characteristics of the models described in Part I and Secs. II B 1–II B 4, and they are not due to the fortuitous choice of a particular set of parameters.

We did not attempt to fit the experimental data quantitatively, since (as pointed out in Part I) it was not possible to convert the measured absorption coefficient values into defect concentrations.

IV. DISCUSSION

Our experimental findings reported in Part I give us three criteria which have to be met by the curves shown in Figs. 1–3. As mentioned in Sec. I we found that both $[V_2]$ and [VO] increased linearly with increasing interstitial oxygen concentration and $[V_2]$ vs [VO] was also linear. A look at Figs. 1–3 shows that only the numerical results of Brelot's model compare favorably with these results. Only this model reproduces qualitatively the three linear relationships found by our experiments. The other models are in qualitative disagreement with our experimental data. On the basis of this we believe that the formation of $O_I I$ pairs is the cause for the increase in the divacancy introduction rate observed for increasing interstitial oxygen concentrations.

This conclusion is consistent with the annealing behavior of the O_iI pair, since the available experimental evidence indicates that the O_iI pair has the thermal stability required for a room-temperature irradiation.[22] Although the O_iI-associated IR band at 935 cm^{-1} is largest after low-temperature (~77 K) irradiation and decreases upon annealing to 310 K it is still present even after annealing at 350 K.[22] We therefore conclude that O_i-I interaction can take place during room temperature electron irradiation and that the resulting complex can be stable at room temperature.

Further support for the correctness of our conclusion comes from the following consideration. In Brelot's model the divacancy increase in oxygen containing silicon is not related to the chemical nature of the oxygen impurity, but only to the fact that oxygen functions as a Si self-interstitial trap. Therefore replacing oxygen by any other interstitial trap should effect a similar divacancy increase. This actually has been observed for carbon.[23,24] Alternatively we could directly vary the steady-state vacancy concentration by changing the concentration of vacancy sinks (impurities, surfaces, etc.), and we should observe a divacancy change. Such an experiment has been done by Wang *et al.*[25] They studied the surface-defect distributions of electron-irradiated silicon by transient capacitance techniques. They observed a depletion of defects near the surface and attributed it to vacancy trapping at the surface. further they found that both VO and V_2 had the same spatial dependence. We reanalyzed their data for the divacancy and the VO center. Plotting $[V_2]$ vs [VO] gives the graph shown in Fig. 4. We note that $[V_2]$ depends linearly on [VO]. The behavior is in qualitative agreement with the curve of Brelot's model in Fig. 3. This further supports the notion that oxygen can influence the steady-state vacancy concentration by interstitial trapping.

The significance of our experimental result that $[V_2]$ can be proportional to [VO] (i.e., that $[V_2]$ reflects the steady-state single-vacancy concentration) and the subsequent explanation of this relationship by Brelot's model is in its relation to the question of divacancy production *per se*. Since there is conclusive experimental evidence that divacancies can form by the direct collision process during electron irradiation of silicon[26,27] for a long time the formation of divacancies via combination of two vacancies was thought to be negligible. We can now answer the question of what fraction of a certain divacancy concentration introduced by *room-temperature* electron irradiation was produced by a direct collision process and what fraction is due to the agglomeration of single vacancies. In our experiment we found[1]:

$$\alpha_{V_2} = 0.57\alpha_{VO} + 0.25\ \text{cm}^{-1}. \tag{18}$$

Since the fraction of α_{V_2} which is proportional to α_{VO} is due to the combination of two vacancies while the constant gives an upper limit of directly introduced divacancies we can rewrite Eq. (18) as

$$\alpha_{V_2} = \alpha_{\text{indirect}} + \alpha_{\text{direct}} \tag{19}$$

with $\alpha_{\text{indirect}} = 0.57\alpha_{VO}$ and $\alpha_{\text{direct}} = 0.25\ \text{cm}^{-1}$. For the largest α_{VO} we had $\alpha_{V_2} = 1.2\ \text{cm}^{-1}$.[1] We then get $\alpha_{\text{indirect}}:\alpha_{\text{direct}} = 0.95{:}0.25$ and we therefore have that the indirect production rate of divacancies can be four times as large as the direct production rate during room-temperature irradiation with 2-MeV electrons. It is interesting that Barnes found a similar ratio in a low-temperature (76 K) neutron-damage experiment. Barnes observed immediately after irradiation at 76 K (i.e., with the single vacancies immobile) only 25% of the maximum $[V_2]$ value which was obtained after a 330-K anneal.[28] Therefore in his case the indirect production rate of V_2 was three times as large as the direct production rate.

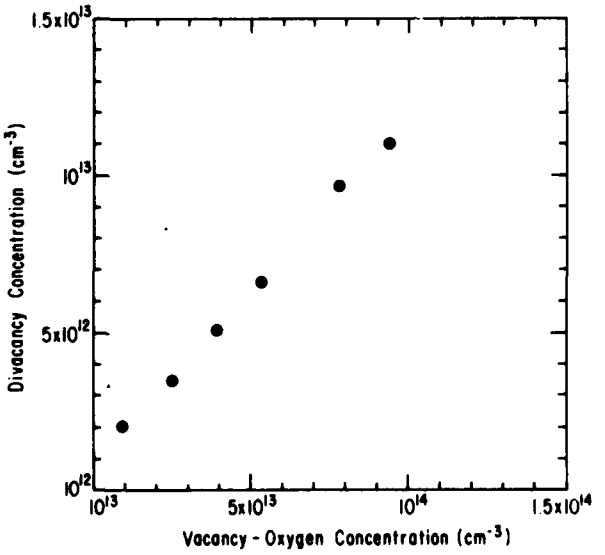

FIG. 4. The data of Wang *et al.*,[25] but here plotting divacancy concentration ($E_c - 0.25$ eV energy level) vs vacancy-oxygen concentration ($E_c - 0.18$ eV energy level).

V. SUMMARY

Numerical tests of possible models for the oxygen dependence of the divacancy introduction rate in at room-temperature electron-irradiated silicon were performed on a computer. Only one of the models can reproduce all the experimental results.[1] The other models give results which are in qualitative disagreement with the experimental data. We envision the mechanism responsible for the enhancement of divacancy production as follows: Interstitial oxygen prevents by the capture of Si self-interstitials interstitial-vacancy recombination. This in turn increases the steady-state vacancy concentration and ultimately enhances the divacancy concentration via agglomeration of two single vacancies.

Our experimental and modeling results show that the indirect formation of divacancies during room-temperature irradiation of silicon with 2-MeV electrons can be more important than the production of V_2 by the single collision process.

ACKNOWLEDGMENT

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